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(71) Applicant: **Abion Corporation Co. Ltd.  
Tokyo 158 (JP)**

(72) Inventor: **Ohtsuka, Riichiro  
Yamato-gun, Fukuoka-ken 835 (JP)**

(74) Representative: **Allard, Susan Joyce et al  
BOULT WADE TENNANT,  
27 Furnival Street  
London EC4A 1PQ (GB)**

(54) **A method of protecting agricultural products**

(57) A method of protecting agricultural products in a facility for the cultivation thereof by exterminating microbes, sterilizing and protecting against insect pests

and of adding an aroma to the facility, which comprises heating in the facility a composition comprising a volatile food additive.

**Description****BACKGROUND OF THE INVENTION**5    **1. Field of the Invention**

This invention relates to a method of protecting plants or crops in a facility or installation for cultivating agricultural products (which will hereinafter be referred to as "facility plants") from insect pests, microorganisms and pathogenic microbes, a method of imparting aroma thereto in a facility and a composition used for these methods. More particularly, 10 it is concerned with a method of protecting facility plants from insect pests, decreasing bad influences on the human body, economizing working labors and destroying offensive order due to organic substances and agricultural chemicals through aromatic effect to add tranquilization by evaporating and diffusing a high safety composition comprising, as a predominant component, volatile food additive such as cinnamic aldehyde or a volatile material including agricultural chemicals in a facility using a warming air boiler or electric heating plate for agriculture.

15    **2. Description of the Prior Art**

As a countermeasure for protecting plants from insect pests or pathogenic microbes, there have been proposed methods comprising diluting agricultural chemicals or plant protectors or activators with water and then spraying them 20 on plants by means of power sprayers or fogging machines, methods comprising spraying or spreading powder materials such as flow dust agents and smoking methods comprising using smoking agents, but these methods have both merits and demerits as to efficiency, safety, working property, investment cost, etc.

For example, as the method comprising spraying an emulsion containing an agricultural chemical on an agricultural 25 products by means of a power sprayer, it is generally carried out to dilute an agricultural chemical (insecticides, microbicides) with water in a suitable concentration, to spray the resulting emulsion on a plant, thus directly reacting it with insect pests, and to protect the plant.

Use of an emulsion containing cinnamic aldehyde for the protection of plants or crops is disclosed in Japanese Patent Publication No. 32283/1986 and US Patent No. 4,978,686.

It is generally said that agricultural products in facilities tend to be attacked by pathogenic microbes or insect pests 30 depending on cultivating ambient conditions such as temperature, humidity, light, etc. and conditions for improving plants. Thus, extermination of pathogenic microbes or insect pests has been carried out by treating agricultural chemicals or plant activators, but in the prior art methods, such an extermination cannot be said sufficient with respect to applied labors, investment costs, safety, extermination effects and bad smell prevention.

35    **SUMMARY OF THE INVENTION**

It is an object of the present invention to provide a method for protecting agricultural products or plants, whereby amounts of agricultural chemicals to be used can be reduced, influences upon environment, agricultural chemicals and human bodies can be decreased and applied labors for the treatment and investment costs can be decreased.

40    It is another object of the present invention to provide an emulsion composition for protecting agricultural products or plants, which is effective as an aromatic agent in a facility for cultivating agricultural products, having a high safety and characteristic odor, because its predominant component is a food additive and perfume, and further is also sufficiently effective as a countermeasure for offensive odor.

45    These objects can be attained by a method of protecting agricultural products comprising heating a composition containing a volatile food additive in a facility for cultivating agricultural products, thereby evaporating and diffusing in the facility, and effecting extermination of microbes, sterilization, protection from insect pests and addition of aroma in the facility.

**DETAILED DESCRIPTION OF THE INVENTION**

50    The present invention has been made so as to solve the above described problems of the prior art and the feature of the present invention consists in maintaining low density of plant insect pests and suppressing development of insect pests in easy manner by treating a composition containing a volatile food additive, typical of which is cinnamic aldehyde having sufficient estimation of safety, utilizing a warming air boiler, electric heating plate, electric or oil stove or humidifier of heating type for facility plants, having already been installed in a facility.

The above described objects of the present invention can be attained by the following methods and compositions according to the present invention and preferred embodiments thereof.

(1) A method of protecting agricultural products comprising heating a composition containing a volatile food additive in a facility for cultivating agricultural products, thereby evaporating and diffusing the volatile food additive in the facility, and effecting extermination of microbes, sterilization, protection from insect pests and addition of aroma in the facility.

5 (2) The method of protecting agricultural products, as described in the above (1), wherein the composition is an emulsion containing an emulsifier and at least one member selected from the group consisting of cinnamic aldehyde, Hinokitiol (extract), allylmustard oils and garlic extracts.

10 (3) The method of protecting agricultural products, as described in the above (1), wherein the heating is carried out by utilizing a warming air boiler electric heating plate electric or oil stove or humidifier of heating typef or agriculture.

15 (4) The method of protecting agricultural products, as described in any one of the above (1) to (3), wherein the volatile food additive has a concentration of 0.05 to 0.5 ppm in the air in a facility for cultivating agricultural products.

(5) An emulsion composition for protecting agricultural products in a facility for cultivating agricultural products, comprising an emulsifier and at least one member selected from the group consisting of cinnamic aldehyde, Hinokitiol(extract), allylmustard oils and garlic extracts.

20 (6) An emulsion composition for protecting agricultural products in a facility for cultivating agricultural products, comprising an emulsifier and at least two members selected from the group consisting of cinnamic aldehyde, Hinokitiol(extract), allylmustard oils and garlic extracts, the cinnamic aldehyde being contained as an essential component.

25 As a composition containing at least one of volatile food additives used in the above described method (1), there is used one containing at least one of cinnamic aldehyde (nonpoisonous material represented by chemical formula C<sub>9</sub>H<sub>8</sub>O and disclosed in an official document as a perfume and food additive), Hinokitiol (food additive represented by molecular formula C<sub>10</sub>H<sub>12</sub>O<sub>2</sub> and contained in a large amount in a part of wood belonging to Hinoki family or in Hinoki hiba arborvitae of hiba arborvitae genus in Japan), allylmustard oils (food additive represented by molecular formula C<sub>4</sub>H<sub>5</sub>NS and contained as a predominant component in wasabi oils or mustard oils) and garlic extracts. In particular, it is preferable to use cinnamic aldehyde as an essential component and other components as optional components.

30 In the above described method (3), the above described composition is converted into an emulsion diluted with water or a solvent, arranged in the vicinity of an inner warm air blow-off port of a warming air boiler provided in a facility for cultivating agricultural products, for example, a vinyl plastic hothouse or plastic film house and then evaporated, diffused and uniformly spread in the facility by warm air of the boiler. The temperature near the warm air blow-off port is ordinarily 20 to 50 °C, preferably about 40 to 43 °C and the concentration of volatile components in the air is generally 0.05 to 0.5 ppm, preferably 0.1 to 0.3 ppm.

35 The electric heating plate is composed of a number of plates for heating to accelerate evaporation of the volatile components and to utilize natural convection in the facility and thereby spreading the food additive in the facility. The above desctribed concentration of the volatile components in the air can be realized by controlling the number of plates and the temperature.

40 The emulsion composition for protecting agricultural products, described in the above described (5) and (6), is preferably used in the form of an emulsion of oil-in-water type comprising generally at least one of cinnamic aldehyde, Hinokitiol (extract) (main component:tuyapricin), allylmustard oils (another name: allyl isothiocyanate) and garlic extracts (Liliaceae Allium sativum, obtained by extracting bulbs of Allium sativum with an organic solvent). These components can be used, individually or in combination, preferably in a concentration of 15 to 30 weight %, more preferably 20 to 25 weight % in the emulsion. Percents are to be taken as those by weight unless otherwise indicated.

45 For example, 20 % of cinnamic aldehyde, 5 % of polyoxyethylene polyoxypropylene glycol, 5 % of oleic acid and 70 % of water are mixed by stirring at a high rate using a stirrer rotating at 3600 to 1500 rpm to obtain an emulsion of oil-in-water type. The thus obtained emulsion is suitably diluted with water (ordinarily about 10 to 20 times) to hold antimicrobial acitivity to various plant microbes and protection effect from insect pests.

50 Of the above described food additives, cinnamic aldehyde is a compound having a molecular formula of C<sub>9</sub>H<sub>8</sub>O, molecular weight of 132.16, a specific gravity of 1.056, boiling point of 252°C (at 760 mmHg, decomposed in part) and 128-130 °C (at 20 mmHg) and melting point of -7 °C, that is a transparent colorless to light yellow liquid and has cinnamon perfume. This is designated as a food additive and has widely been used as perfumes for foods, spices and medicines. Hinokitiol is a white to light yellow crystal or crystalline powder which has peculiar oder and a molecular formula of C<sub>10</sub>H<sub>12</sub>O<sub>2</sub>, molecular weight of 164.20 and melting point of 48 to 52 °C. Its extracted product is printed as a preservatives in a List of Food Additives in addition to chemical synthetic materials and has been used as a food use. In the present invention, the extracted product is preferably used after being dissolved in an alcohol, in particular, ethanol in an amount of 5 to 10 times by weight as much as it.

55 Allylmustard oil (allyl isothiocyanate) is a transparent white to light yellow oily liquid having a molecular formula of C<sub>4</sub>H<sub>5</sub>NS, molecular weight of 99.16, specific gravity of 1.018 to 1.023 and boiling point of 148 to 154 °C, that has

peculiar stimulative oder. This is a predominant component in an essential oil of wasabi or mustard, which has been designated as a food additive and has mainly been used as a spice or medicine. Garlic extracts can be obtained in the form of an oil, paste, powder, etc. by extracting bulbs of Allium sativum with an organic solvent and printed as a preparation agent in a List of Food Additives in addition to chemical synthetic materials.

5 For emulsifying the above described food additive, it is preferable to use polyoxyethylene polyoxypropylene glycol or oleic acid as an emulsifier.

The present invention will be illustrated by the following Examples in detail without limiting the same.

#### Example 1

10 A mixture of 20 % of cinnamic aldehyde, 10 % of an emulsifier (5 % of polyoxyethylene polyoxypropylene glycol, 5 % of oleic acid) and 70 % of water was stirred and emulsified for 5 minutes by means of a high speed stirrer. (A).

A mixture of 19 % of cinnamic aldehyde, 0.1 % of Hinokitiol (extract), 1 % of garlic extracts, 10 % of the above described emulsifier and 69.9 % of water was stirred and emulsified for 5 minutes by means of a high speed stirrer. (B).

15 A mixture of 18 % of cinnamic aldehyde, 1 % of allylmustard oil (allyl isothiocyanate), 1 % of garlic extract, 10 % of the above described emulsifier and 70 % of water was stirred and emulsified for 5 minutes by means of a high speed stirrer. (C).

Food additive emulsions prepared by the above described three kinds of production processes exhibited high stability and good suspensibility even if diluted with water (20 times) and furthermore, its solution had peculiar aroma.

20 In order to investigate the antimicrobial acitivity to various plant microbes, the following tests were carried out.

Previously cultured plant microbes (*Botrytis cinerea*, *Phytophthora capsici*) were cut in 5 mm square and implanted on PSA culture medium in a petri. Furthre, the emulsions A, B and C were dropwise added to the scale, covered and cultured at 25 °C for 5 days, after which growth state of the microbes was investigated. The results are shown in Table 1, from which antimicrobial activity can be found as to each of the emulsions. After 5 days, growth of the microbes proceeded in Control Section, while growth of the microbes was hardly confirmed or considerably suppressed in Sections A, B and C.

Table 1

Antimicrobial Activity Effect of Plant Microbes by Food Additive Emulsion		
	Growth State of Plant Microbes After 5 Days	
	<i>Botrytis cinerea</i>	<i>Phytophthora capsici</i>
Section A	±	-
Section B	+	±
Section C	-	-
Control Section	+++	+++

#### Example 2

40 Using the food additive emulsions (A), (B) and (C), prepared in the similar manner to Example 1, the following tests were carried out in order to investigate damage of plants from agricultural chemicals and influences upon the human body and to know such a concentration as not giving offensive smell next morning.

45 A solution obtained by adding a predetermined amount of each of the emulsions to 1000 ml of water was placed near an air blow-off port in a warming air boiler for seven days and nights and evaporated and filled in a vinyl plastic hothouse by operating the boiler. The temperature in the vicinity of the blow-off port was 40 to 43 °C and the concentration of volatile components was 0.03 to 0.5 ppm.

As is evident from results shown in Table 2, there were found no damage of plants from agricultural chemicals, nor bad influences upon the human body when using at most 100 ml of each of the emulsions per 20 a of the vinyl plastic hothouse.

Table 2

Damage of Plants from Agricultural Chemicals and Degree of Smell by Food Additive Emulsion in Vinyl Plastic Hothouse						
		Tomato	Eggplant	Cucumber	Chrysanthemum	Orange
<b>Emulsion A</b>						
20 ml Damage	-	-	-	-	-	-
Smell	very little	very little				
50 ml Damage	-	-	-	-	-	-
Smell	little	little	little	little	little	little
100 ml Damage	-	-	-	-	-	-
Smell	medium	medium	medium	medium	medium	medium
200 ml Damage	±	-	-	-	±	-
Smell	much	much	much	much	much	much
<b>Emulsion B</b>						
20 ml Damage	-	-	-	-	-	-
Smell	very little	very little				
50 ml Damage	-	-	-	-	-	-
Smell	little	little	little	little	little	little
100 ml Damage	-	-	-	-	-	-
Smell	medium	medium	medium	medium	medium	medium
200 ml Damage	±	-	-	±	±	-
Smell	much	much	much	much	much	much
<b>Emulsion C</b>						
20 ml Damage	-	-	-	-	-	-
Smell	very little	very little				
50 ml Damage	-	-	-	-	-	-
Smell	little	little	little	little	little	little
100 ml Damage	-	-	-	-	-	-
Smell	medium	medium	medium	medium	medium	medium
200 ml Damage	-	±	±	±	+	±
Smell	much	much	much	much	much	much
Note: (-) means no damage.						

## Example 3

Using the food additive emulsions (A), (B) and (C), prepared in the similar manner to Example 1, the following tests were carried out in order to investigate degrees of disease by main microbes and degrees of development of main insect pests in each plant.

A solution was prepared by adding a predetermined amount of each of the emulsions to 1000 ml of water and placed near an air blow-off port in a warming air boiler for 30 days and evaporated and filled in a vinyl plastic hothouse by operating the boiler in an analogous manner to Example 2. The temperature in the vicinity of the blow-off port was 35 to 43 °C and the concentration of volatile components was 0.03 to 0.2 ppm.

For comparison, the plants were treated with agricultural chemicals (microbicide, insecticide) two times at an interval of 10 days with provision of a non-treated section.

As is evident from results shown in Table 3, there was found decreased disease or development of main insect pests when using each of the emulsions in a range of 50 to 100 ml per 20 a of a vinyl plastic hothouse, which effect was substantially similar to the extermination effect by agricultural chemicals. In particular, an effect for main pests was markedly found.

Table 3

Extermination Effect of Main Plant Insect Pests by Food Additive Emulsions in Facility						
	Tomato		Cucumber		Orange	
	Leaf Mold*	Greenhouse White Fly**	Downy Mildew*	Aphid**	Gray Mold*	Spider Mite**
Emulsion A						
20 ml	93.0	98.8	95.8	101.2	99.7	99.6
50 ml	44.6	85.4	30.8	80.1	50.5	87.6
100 ml	36.2	79.0	27.1	70.3	30.2	66.8
Emulsion B						
20 ml	88.7	95.7	93.3	70.6	98.8	92.1
50 ml	41.8	79.0	50.4	44.3	55.0	60.9
100 ml	38.9	60.1	33.3	10.5	47.0	55.7
Emulsion C						
20 ml	84.8	92.9	87.9	69.9	80.5	90.5
50 ml	33.3	75.5	43.4	30.8	49.1	81.0
100 ml	29.5	57.2	39.0	25.4	28.3	60.3
Control Section	30.3	34.3	28.3	25.6	56.3	80.7
Non-treated Section	100.0	100.0	100.0	100.0	100.0	100.0

Note: \*) disease index

\*\*) development index

Disease index and Development index when index of Non-treated

Section is 100.0.

### Advantages of the Invention

As apparent from the foregoing illustrations and results of Examples, the present invention provides a method of protecting plants or crops in a facility or installation for cultivating agricultural products from insect pests, microorganisms and pathogenic microbes and a method of imparting aroma thereto in a facility, wherein agricultural plants are protected from insect pests and pathogenic microbes without unfavorably affecting the agricultural plants and labor-saving in the extermination operation and safety on the human body and agricultural products are improved by using a composition containing a volatile food additive, typical of which is cinnamic aldehyde having sufficient estimation of safety through a warming air boiler or electric heating plate for facility plants, having already been installed in a facility.

### Claims

1. A method of protecting agricultural products in a facility for the cultivation thereof by exterminating microbes, sterilizing and protecting against insect pests and of adding an aroma to the facility, which comprises heating in the facility a composition comprising a volatile food additive.
2. The method as claimed in Claim 1 wherein the composition is an emulsion containing an emulsifier and at least one member selected from cinnamic aldehyde, Hinokitiol, allylmustard oils and garlic extracts.
3. The method as claimed in Claim 1 or 2 wherein the heating is carried out by a warming air boiler or an electric heating plate for agriculture.
4. The method as claimed in any one of Claims 1 to 3 wherein the volatile food additive has a concentration of 0.05 to 0.5 ppm in the air in the facility.

5. An emulsion composition for protecting agricultural products in a facility for the cultivation thereof, comprising an emulsifier and at least one member selected from cinnamic aldehyde, Hinokitiol, allylmustard oils and garlic extracts.
- 5      6. An emulsion composition according to Claim 5 which comprises cinnamic aldehyde and at least one member selected from Hinokitiol, allylmustard oils and garlic extracts.

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## EUROPEAN SEARCH REPORT

Application Number  
EP 97 30 9340

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Place of search	Date of completion of the search	Examiner	
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CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			



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